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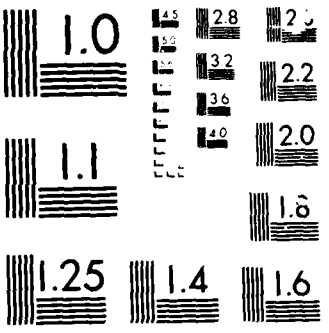
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Theoretical techniques have been developed to describe a variety of laser-induced molecular rate processes occurring at solid surfaces which are involved in heterogeneous catalysis. Such processes include adsorption, migration, chemical reactions and desorption. The role of surface phonons in laser-selective processes and laser heating has been analyzed. The importance of electronic degrees of freedom has been considered for semiconductor and metal substrates, with special emphasis on the laser excitation of surface states. Surface-modified photochemistry has also been investigated, where the effect of a metal surface on the resonance fluorescence spectrum of a laser-driven atom/molecule has been assessed by means of surface-dressed optical Bloch equations. It is seen that the spectrum can be significantly different from the gas-phase case. Two related gas-surface collision processes have also been studied. First, the feasibility of the formation of electron-hole pairs in a semiconductor by vibrationally-excited molecules has been explored. Second, charge transfer in ion-surface collisions

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19. (continued) has been examined for both one-electron and two-electron transfer processes. The former corresponds to ion neutralization and has been looked at for proton neutralization at alkali-halide surfaces (insulators). The latter corresponds to negative-ion formation and has been considered for metal substrates both cold and at finite temperatures (up to 3000 K). Finally, work has been initiated on microstructures and rough structures, including clusters and surface gratings.

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CONTENTS

#1

Summary

Theoretical techniques have been developed to describe a variety of laser-induced molecular rate processes occurring at solid surfaces which are involved in heterogeneous catalysis. Such processes include adsorption, migration, chemical reactions and desorption. The role of surface phonons in laser-selective processes and laser heating has been analyzed. The importance of electronic degrees of freedom has been considered for semiconductor and metal substrates, with special emphasis on the laser excitation of surface states. Surface-modified photochemistry has also been investigated, where the effect of a metal surface on the resonance fluorescence spectrum of a laser-driven atom/molecule has been assessed by means of surface-dressed optical Bloch equations. It is seen that the spectrum can be significantly different from the gas-phase case. Two related gas-surface collision processes have also been studied. First, the feasibility of the formation of electron-hole pairs in a semiconductor by vibrationally-excited molecules has been explored. Second, charge transfer in ion-surface collisions has been examined for both one-electron and two-electron transfer processes. The former corresponds to ion neutralization and has been looked at for proton neutralization at alkali-halide surfaces (insulators). The latter corresponds to negative-ion formation and has been considered for metal substrates, both cold and at finite temperatures (up to 3000 K). Finally, work has been initiated on microstructures and rough structures, including clusters and surface gratings.

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Publications

Each manuscript listed below corresponds by number to the Technical Report previously submitted to the Office of Naval Research for Contract N00014-80-C-0472. The major portion of these are journal articles, where the remainder are book chapters and conference proceedings. The one exception is #8, which is the Ph.D. thesis of Dr. Jui-teng Lin. Another Ph.D. thesis in chemistry supported by ONR but not submitted as a Technical Report is "Charge Transfer and Electronic Relaxation in Ion-Surface Scattering" by Dr. Franco Battaglia (University of Rochester, 1985). The results of this thesis are contained in #40, 55, 63, and 66 listed below.

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